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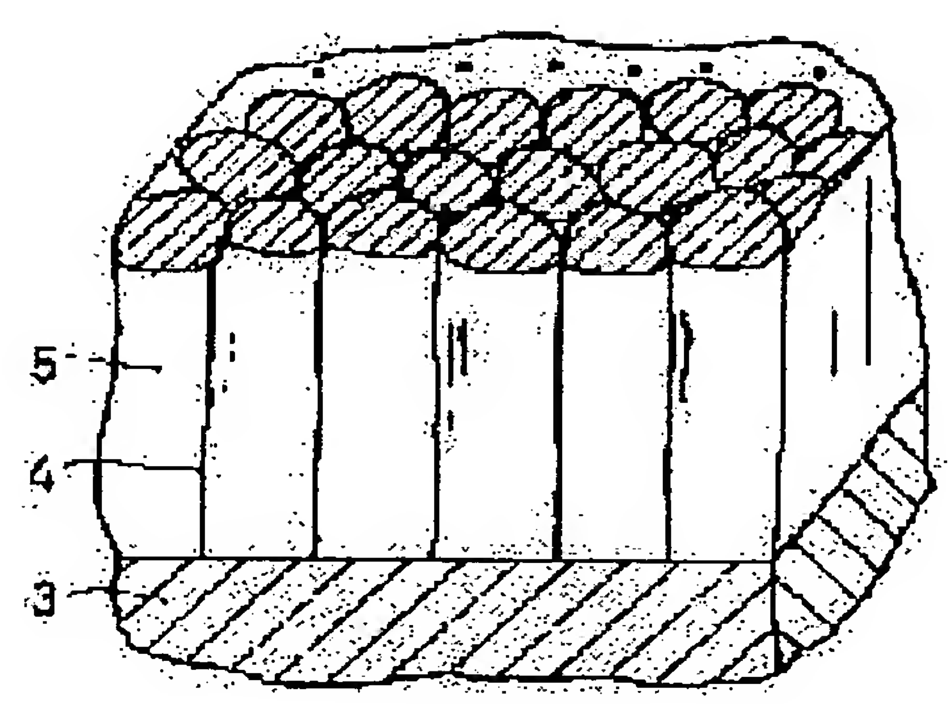
(54) RARE EARTH ELEMENT-FE-B-THIN FILM PERMANENT MAGNET

(57)Abstract:
PURPOSE: To get excellent magnetic property even if a permanent magnet is thin by putting this magnet in pillar-shaped structure where a grain boundary being a nonmagnetizing phase made through in the direction of film thickness and the magnetic main phases separated among particle phases extend in the direction of film thickness, and putting the aspect rate of the pillar-shaped structure to a specified value or over.
CONSTITUTION: A rare earth element-Fe-B thin film permanent magnet, which consists of alloy having R (R shows Nd and/or Pr out of rare earth elements), Fe, and B for its main ingredients, is made in pillar-shaped structure where a grain boundary 4 being a nonmagnetizing phase is made through in the direction of film thickness, and also magnetic main phases 5 magnetically separated among particles extend in the direction of thickness, and besides the aspect ratio (length/diameter ratio) is 5 or over. And, this has excellent magnetic property even if it is thin. Hereby, a rare earth element-Fe-B thin film permanent magnet, which can exhibit favorable magnetic property besides being thin, can be materialized.



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CLAIMS

[Claim(s)]
[Claim 1] While the grain boundary which is the rare-earth-elements-Fe-B system thin film permanent magnet which consists of an alloy which uses R (R expresses Nd and/or Pr among rare earth elements), and Fe and B as a principal component, and is a nonmagnetic phase is pierced through and formed in the direction of thickness The rare-earth-elements-Fe-B system thin film permanent magnet which has the columnar structure to which the magnetic main phase separated magnetically extends in the direction of thickness, and is formed between particles, and is characterized by the aspect ratio (die length / diameter ratio) of this columnar structure phase being five or more.
[Claim 2] Atom [R:8 to 18] %, atom [B:5 to 10] %, the remainder: The rare-earth-elements-Fe-B system thin film permanent magnet according to claim 1 which is what consists of Fe substantially.
[Claim 3] The rare-earth-elements-Fe-B system thin film permanent magnet according to claim 2 which permutes a part of Fe by Ag below 1.5 atom %.

[Translation done.]

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DETAILED DESCRIPTION

[Detailed Description of the Invention]
[0001]
[Industrial Application] This magnet makes the start various electric products and a computer

for home use about the rare-earth-elements-Fe-B system thin film permanent magnet which has outstanding magnetic properties, and this invention is useful as a material of the electrical and electric equipment and electronic ingredients, such as a high performance small motor.
[0002]
[Description of the Prior Art] A permanent magnet is an ingredient which generates a field, even if it does not supply electric energy from the exterior, and the need makes a subject the above electrical and electric equipment and electronic ingredients, and has been growing increasingly. In recent years, the rare earth magnet attracts attention as the 3rd permanent magnet which ranks second to a ferrite magnet and an alnico magnet. This rare earth magnet was expected to demonstrate the outstanding magnetic property which can contribute to a miniaturization and highly-precise-izing of an electric product or a precision equipment, and shows progress with active physical-properties research side and industrial-engineering side. Permanent magnets, such as a rare-earth-elements-transition element-B system, for example, Nd-Fe-B, and Pr-Fe-B, are expected especially especially in recent years, and the maximum energy product [it may express with (BH) max below] shown by the product of a residual magnetic flux density (Br) and coercive force (iHc) is said to be high while the permanent magnet of this system has high coercive force (iHc).
[0003] Although the permanent magnet presentation of this invention makes a fundamental component rare-earth-elements (however, Nd and/or Pr)-Fe-B and also explains the evaluation in full detail later on including Ag as occasion demands as the 4th component Suppose for convenience that 3 element magnet of a rare-earth-elements-Fe-B system (it may be hereafter called a R-Fe-B system for short) is taken up typically, and is described in the following explanation.
[0004] By the way, as the manufacture approach of a R-Fe-B system magnet, the sintering process, the melt quenching method, etc. are known as a typical approach, and formation of a thin film permanent magnet is performed by these approaches. It is known that the magnetic properties of a magnet ingredient will be greatly influenced in a detailed organization. And with a sintering mold magnet By considering as the structure where the grain boundary phase 1 fully encloses the main phase crystal grain 2 as shown in drawing 2 , (Three in **** shows a substrate) The generating site of a reverse magnetic domain is decreased and it is said that magnetic properties can be raised (the Japan Institute of Metals, and 57th volume No. 4 (1993) P 470-477). Moreover, with the high-speed quenching mold magnet, it consists of detailed main phase crystal grain and amorphous substances, and it is said by adjusting a quenching rate that high magnetic properties are acquired (the Institute of Electrical Engineers of Japan paper magazine A, 113 No. 4 (1993), P251-260).
[0005]
[Problem(s) to be Solved by the Invention] However, when it sees in the direction of thickness in the R-Fe-B system permanent magnet obtained by a sintering process which was described above, and liquid sudden cooling, crystal grain is structure irregularly piled up on both sides of the grain boundary phase, and the contribution of magnetic shape anisotropy to a perpendicular magnetic anisotropy is small. Moreover, coercive force is a reverse magnetic-domain type of seasonal prevalence influenced by the generating field of a reverse magnetic domain, and actual coercive force is as small as about 1 of the flux reversal field Ha of the single domain particle which has uniaxial anisotropy theoretically / ten to 2/5. the above from such a thing -- the actual condition is that implementation of the permanent magnet for products which cannot realize the permanent magnet with which it can be satisfied of both the properties of thinning and high-performance-izing by any approach, but can meet enough a demand in recent years called a miniaturization and high-performance-izing is desired.
[0006] On the other hand, the approach by the sputtering method which is looked at by JP,4-99010,A is also proposed as the new formation approach of a R-Fe-B system thin film permanent magnet. This technique grows up C shaft which is an easy axis of the R2 Fe14 B phase which is the main phase in the direction of thickness, and forms the film which has an anisotropy strong against the direction of thickness. However, with this technique, about a nonmagnetic grain boundary phase or magnetic shape anisotropy, it is not taken into

consideration at all, but the further improvement in magnetic properties is desired.
[0007] This invention is made under such a situation, the purpose realizes film with which the effectiveness of magnetic shape anisotropy is given, and a grain boundary phase which serves as a rotation magnetization mold from the conventional reverse magnetic-domain type of seasonal prevalence in a coercive force device is to offer the rare-earth-elements-Fe-B system thin film permanent magnet which is thin meat and can moreover demonstrate good magnetic properties, while separating between particles magnetically.
[0008]
[Means for Solving the Problem] This invention which could attain the above-mentioned purpose is a rare-earth-elements-Fe-B system thin film permanent magnet which consists of an alloy which uses R (R expresses Nd and/or Pr among rare earth elements), and Fe and B as a principal component. While the grain boundary which is a nonmagnetic phase is pierced through and formed in the direction of thickness, between particles, the magnetic main phase separated magnetically has the columnar structure prolonged in the direction of thickness, and is formed. And it is the rare-earth-elements-Fe-B system thin film permanent magnet which has a summary at the point that the aspect ratio (die length / diameter ratio) of this columnar structure phase is five or more. the rare-earth-elements-Fe-B system thin film permanent magnet of this invention -- R:8 to 18 atom %, B:5 to 10 atom %, and remainder: -- the thing of the chemical entity presentation which consists of Fe substantially is desirable, and permuting a part of Fe by Ag in the range below 1.5 atom % also has it. [effective]
[0009]
[Function] this invention persons repeated examination about the relation of the crystalline-structure structure and magnetic properties especially that thin meat should moreover realize a highly efficient permanent magnet on the basis of the technique proposed so far. Consequently, in what the R2 Fe14 B phase which is the grain boundary phase and the magnetic main phase which are a nonmagnetic phase is presenting specific organization, even if it was thin meat, it turned out that extremely excellent magnetic properties are shown. Namely, as shown in drawing 1 , while the grain boundary 4 which is a nonmagnetic phase is pierced through and formed in the direction of thickness The magnetic main phase 5 (for example, R2 Fe14 B phase) separated between particles has the columnar structure prolonged in the direction of thickness, and is formed. And a header and this invention were completed for (BH) max expressing and ****** (ing) compared with a permanent magnet as the aspect ratio (die length / diameter ratio) of this columnar structure phase showed with the conventional technique in the thing of the predetermined range (in addition, three show a substrate among drawing 1).
[0010] Although not the all could be solved about the reason the above effectiveness was acquired by this invention, probably it can think as follows. That is, by the above organization, since the magnetic main phase has [the aspect ratio] the large columnar structure, while magnetic shape anisotropy arises and the magnetic anisotropy of the direction of thickness becomes large in addition to a crystal magnetic anisotropy, since said magnetic main phase is magnetically separated between particles, the magnetic interaction between particles becomes weaker, and it is thought that coercive force becomes large.
[0011] Next, the alloy presentation which constitutes the R-Fe-B system magnet of this invention is explained. As rare earth elements, it needs to be first referred to as Nd and/or Pr. That is, if that the highest magnetic properties are acquired needs to be Pr and/or Nd as rare earth elements since it is Pr and Nd and other rare earth elements are included, the magnetic properties for which it wishes will not be acquired.
[0012] In the R-Fe-B system magnet of this invention, if there is too little R, R2-Fe14-B (an atomic ratio, for example, Pr2 Fe14B) which is the main phase will become is hard to be formed, and magnetic high performance-ization will not be attained. Considering such a viewpoint, as for the rate of R, it is desirable to carry out to more than 8 atom %. On the other hand, if 18 atom % is exceeded about an upper limit, the excess of R rich phase which is a nonmagnetic phase is caused, this serves as a fall of flux density (Br) etc., and appears, and good magnetic properties cannot be demonstrated. In addition, the more desirable presentation rate of R is the range of 12 - 15 atom %.

[0013] On the other hand, as for B, it is desirable to consider as five to 10 atom %, under by pentatomic %, lack of the rate of the main phase volume arises, and it causes the fall of flux density (Br). It is R2 Fe4 B4 which does not have magnetic properties about an another side upper limit. It is desirable to carry out from a viewpoint of preventing the fall of the coercive force (iHc) by the appearance of a phase, to below 10 atom %. In addition, the more desirable range of B is 7 - 9 atom % extent.
[0014] In the R-Fe-B system magnet of this invention, although it is Fe (namely, Fe and an unescapable impurity) substantially except Above R and B, it is also effective to permute a part of Fe by Ag. That is, whenever [magnetic isolated / between the grain children who add Ag] improves, and (BH) max increases. However, if the addition of Ag is made [many / not much], a nonmagnetic grain boundary phase will increase, and it is (BH) max. Since it falls on the contrary, it should carry out below 1.5 atom %. In addition, the more desirable range of the addition of Ag is below 1 atom %.
[0015] Although it is necessary to make the aspect ratio (die length / diameter ratio) of said columnar structure phase (the magnetic main phase) in the R-Fe-B system thin film magnet of this invention or more into five, it becomes what has the small improvement in the property according [this value] to magnetic-shape-anisotropy grant less than by five.
[0016] In addition, this invention persons are an aspect ratio and (BH) max. And when the relation of thickness is considered, it is checking about the following point. Namely, with a Nd-Fe-B system thin film magnet, at 5 micrometers, thickness is the thing of 5-15 and an aspect ratio is (BH) max. 17 - 18MGOe is attained, an aspect ratio is the thing of 20-50 in 20 micrometers of thickness, and it is (BH) max. 14 - 16MGOe was attained.
[0017] Although what is necessary is just to make it form the magnetic main phase which has a columnar structure by the sputtering method fundamentally as an approach of manufacturing the R-Fe-B system thin film permanent magnet of this invention, if the above detailed organizations can be formed, a vacuum deposition method, a CVD method, etc. are also employable. Moreover, although it does not limit especially about the conditions at the time of manufacturing, especially in order to form the above-mentioned organization, it is desirable to make substrate temperature at the time of manufacture into about 500-700 degrees C. That is, at less than 500 degrees C, the film will be [the substrate temperature at this time] in an amorphous condition, hard magnetism is not acquired, but if it exceeds 700 degrees C, phases other than R2 Fe14B (for example, alpha-Fe equality) will generate, and it is (BH) max. It will decrease. Furthermore, sputtering gas ** at the time of manufacturing the R-Fe-B system thin film permanent magnet of this invention is good to make it higher than the conventional approach (for example, 8x10-3Torr extent) (for example, 15x10-3Torr extent).
[0018] Although an example explains this invention to a detail further below, the following example is not the thing of the property which limits this invention, and each thing marked and done to before and the after-mentioned meaning for a design change is included in the technical range of this invention.
[0019]
[Example]
The alloy target into which the example 1 presentation rate was changed was used, and the Nd-Fe-B alloy thin film magnet of various presentation rates was created by the DC sputtering method (sputtering-gas **: 15x10-3Torr). Substrate temperature was made into 600 degrees C at this time. Moreover, the thickness of a thin film was 20 micrometers. Furthermore, it checked that the obtained thin film was the crystalline structure which showed said drawing 1 by electron microscope observation. About the obtained alloy thin film magnet, it is maximum energy product (BH) max by the oscillating sample mold magnetometer (VSM) respectively. It measured. Maximum energy product (BH) max of the thin film magnet which considers Nd, and B and Fe as a basic presentation at drawing 3 A slash shows the presentation range where a value exceeds 14MGOe(s). In addition, the aspect ratios of the thing exceeding 14MGOe(s) were 20-50.
[0020] The alloy target into which the example 2 presentation rate was changed was used, and the Pr-Fe-B alloy thin film magnet of various presentation rates was created by the DC sputtering method (sputtering-gas **: 15x10-3Torr). Substrate temperature was made into 600

degrees C at this time. Moreover, the thickness of a thin film was 20 micrometers. Furthermore, it checked that the obtained thin film was the crystalline structure which showed said drawing 1 by electron microscope observation. About the obtained alloy thin film magnet, it is (BH) max by VSM respectively. It measured. (BH) max of the thin film magnet which considers Pr, and B and Fe as a basic presentation at drawing 4 A slash shows the presentation range where a value exceeds 14MGOe(s). In addition, the aspect ratios of the thing exceeding 14MGOe(s) were 20-50.

[0021] The presentation of B was uniformly carried out to example 3Nd, the presentation of Fe and Ag of the remainder was changed, and membranes were formed. Drawing 5 is Nd12-Fe83-x-B5-Agx. (BH) max by change of membranous Ag addition (x) It is the graph which shows change. In addition, the result of drawing 5 is performed in each presentation on the membrane formation conditions from which maximum was obtained. Moreover, the obtained alloy thin film magnet checked that an aspect ratio was the crystalline structure which showed drawing 1 by 20-50 by electron microscope observation. Addition of Ag below 1.5 atom % is (BH) max so that clearly from this result. It turns out that it is effective in improvement.

[0022] The presentation of B was uniformly carried out to example 4Pr, the presentation of Fe and Ag of the remainder was changed, and membranes were formed. Drawing 6 is Pr12-Fe83-x-B5-Agx. (BH) max by change of membranous Ag addition (x) It is the graph which shows change. In addition, the result of drawing 6 is performed in each presentation on the membrane formation conditions from which maximum was obtained. Moreover, the obtained alloy thin film magnet checked that an aspect ratio was the crystalline structure which showed drawing 1 by 20-50 by electron microscope observation. Also setting in a Pr-Fe-B system so that clearly from this result, addition of Ag below 1.5 atom % is (BH) max. It turns out that it is effective in improvement.

[0023] The alloy of example of comparison 15.0 atom %Pr-79.2 atom %Fe-5.5 atom %B-0.3 atom %Ag was dissolved from high-frequency heating in the argon gas ambient atmosphere, it blew off on the steel single roll (200mmphi) which rotates by roll surface-velocity 10 m/s, and the quenching thin band was created. The thickness at this time was 20 micrometers. Moreover, it checked that the obtained thin film was the crystalline structure which showed said drawing 2 by electron microscope observation. About the obtained thin film, it is (BH) max by VSM. When measured, only 14MGOe(s) were obtained at the maximum.

[0024]

[Effect of the Invention] This invention is constituted as mentioned above and the rare-earth-elements-Fe-B system thin film permanent magnet which is thin meat and can moreover demonstrate good magnetic properties has been realized.

[Translation done.]

DESCRIPTION OF DRAWINGS

[Brief Description of the Drawings]

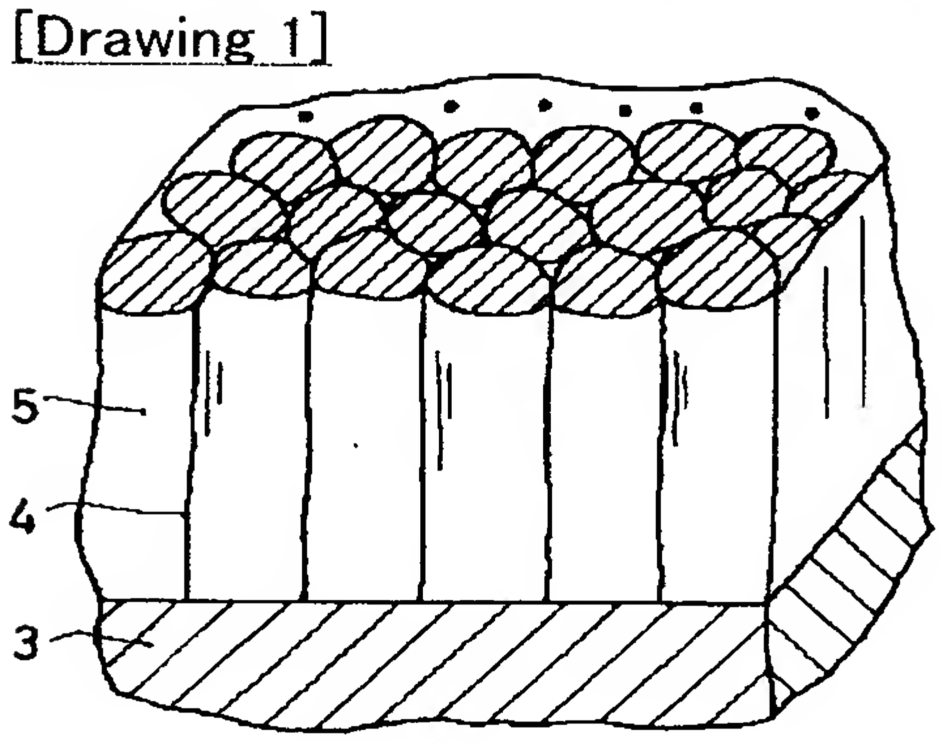
[Drawing 1] It is the mimetic diagram showing the detailed organization of the permanent magnet

concerning this invention.
[Drawing 2] It is the mimetic diagram showing the detailed organization of the conventional permanent magnet.
[Drawing 3] (BH) max of the Nd-Fe-B system thin film magnet of this invention It is the graph which showed the presentation range where a value exceeds 14MGOe(s).
[Drawing 4] (BH) max of the Pr-Fe-B system thin film magnet of this invention It is the graph which showed the presentation range where a value exceeds 14MGOe(s).
[Drawing 5] Nd12-Fe83-x-B5-Agx of this invention (BH) max of a thin film magnet It is the graph which showed change by Ag addition which can be set.
[Drawing 6] Pr12-Fe83-x-B5-Agx of this invention (BH) max of a thin film magnet It is the graph which showed change by Ag addition which can be set.
[Description of Notations]
1 Grain Boundary Phase
2 The Main Phase Crystal Grain
3 Substrate
4 Grain Boundary
5 The Magnetic Main Phase

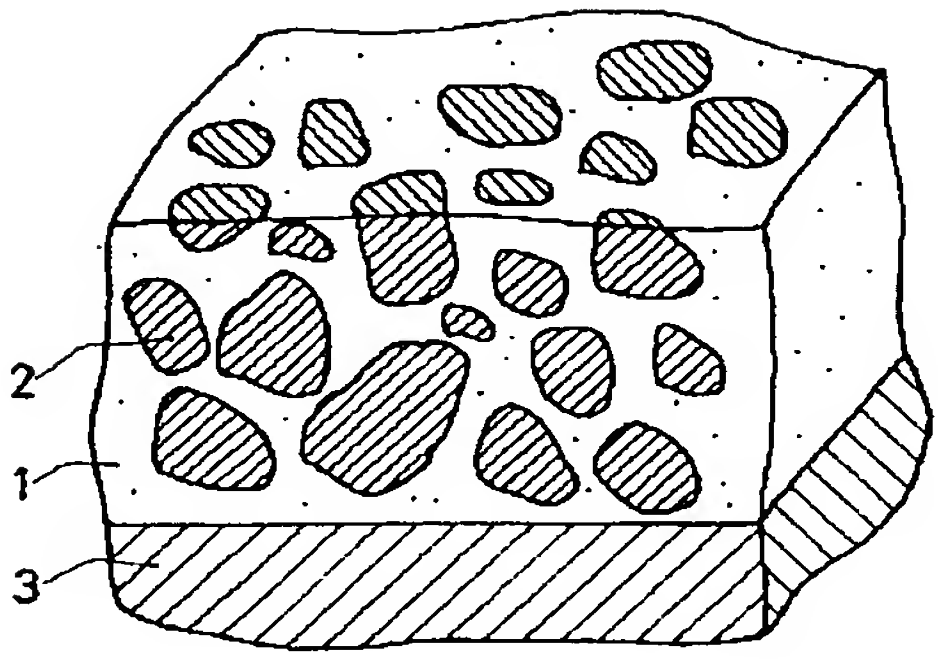
[Translation done.]

DRAWINGS

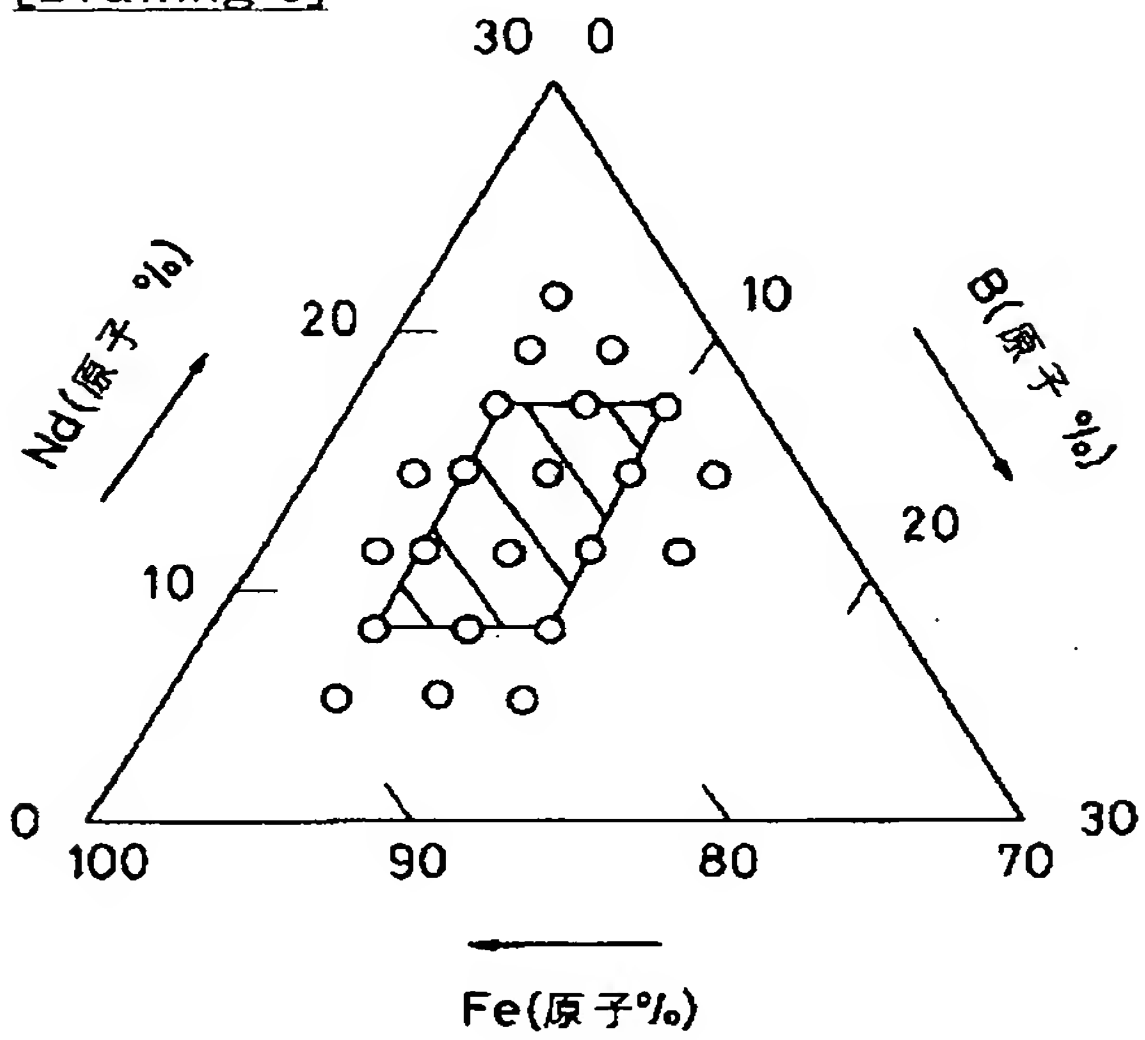
[Drawing 1]



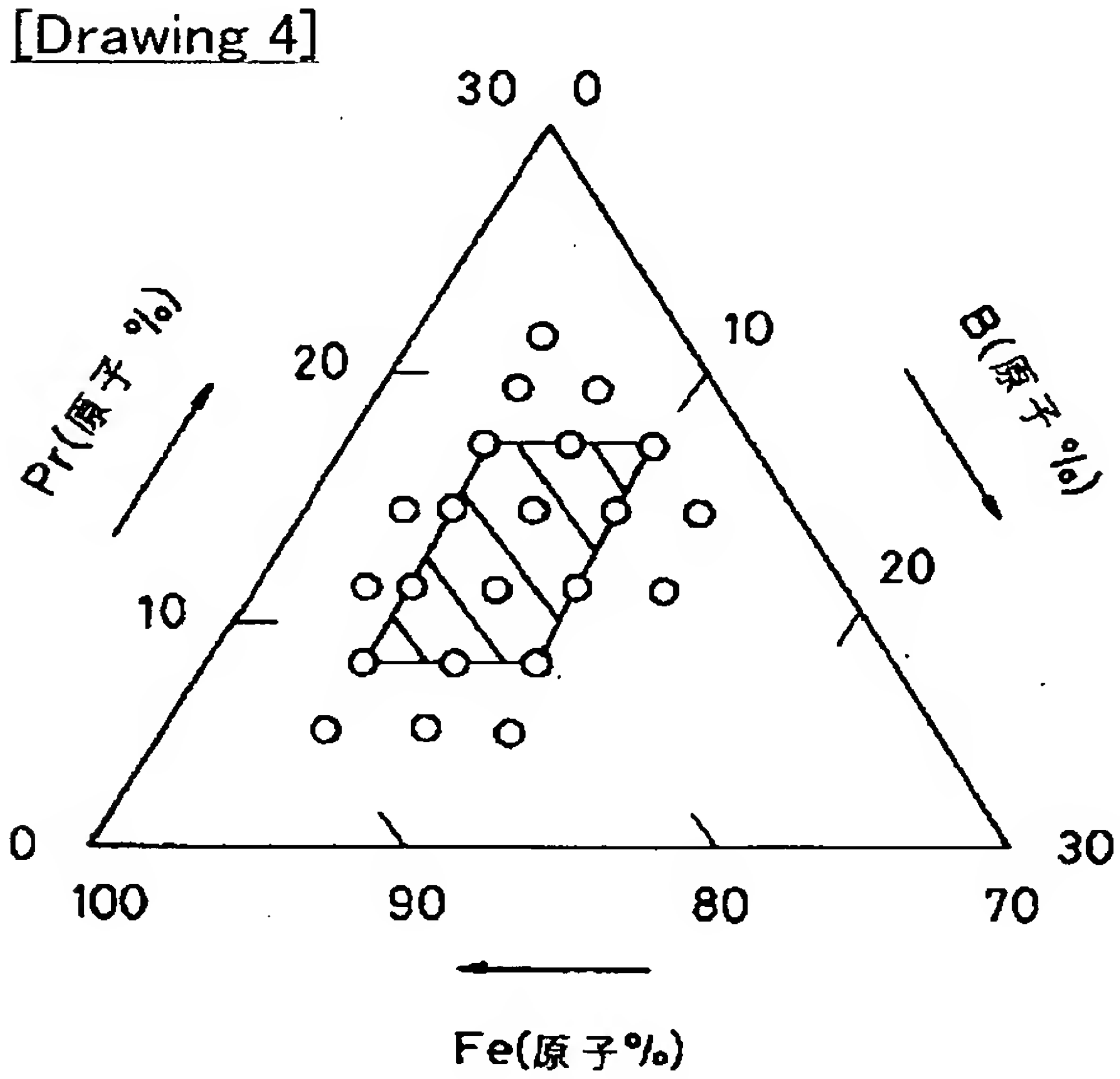
[Drawing 2]



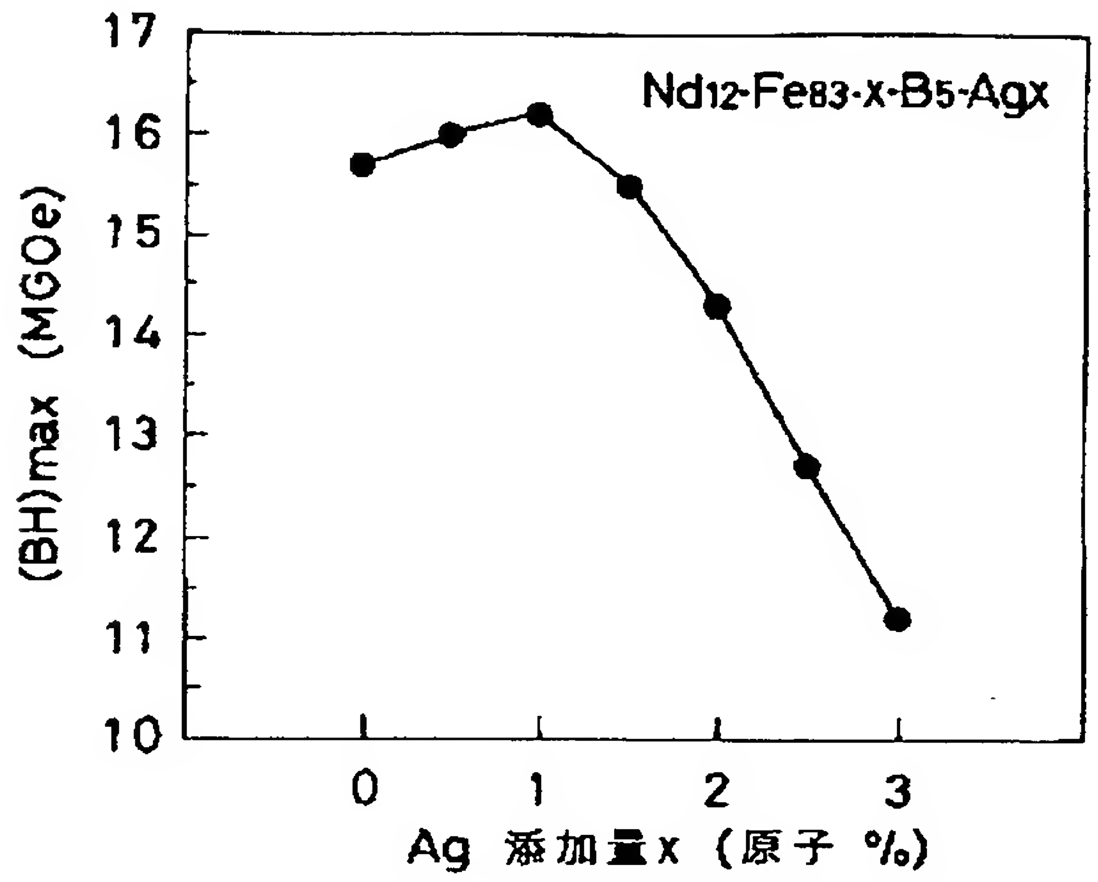
[Drawing 3]



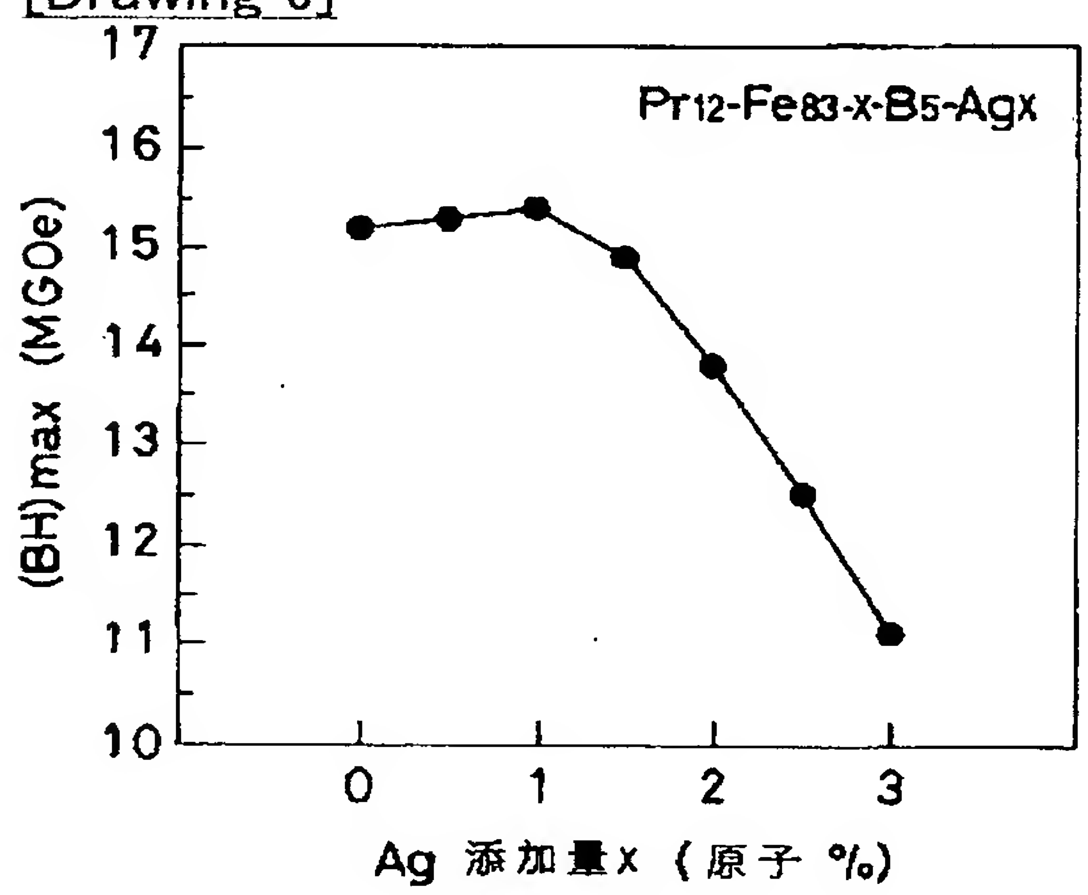
[Drawing 4]



[Drawing 5]



[Drawing 6]



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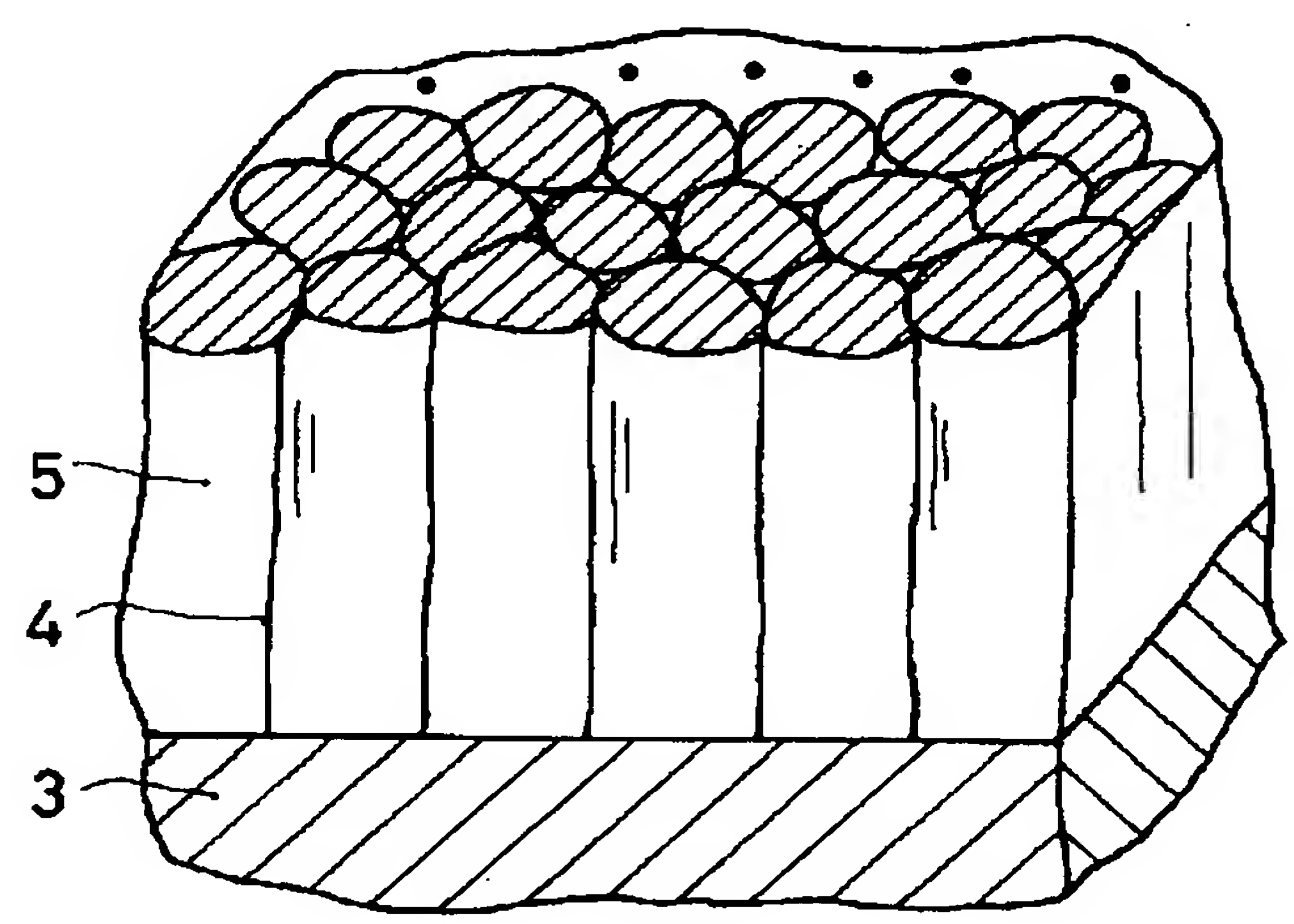
(54)【発明の名称】 希土類元素－Ｆｅ－Ｂ系薄膜永久磁石

(57)【要約】

【目的】 保磁力機構を従来の逆磁区発生型から回転磁化型となるような粒界相が、粒子間を磁気的に分離すると共に、形状磁気異方性の効果が付与される様な膜を実現し、薄肉であってしかも良好な磁気特性を発揮することのできる希土類元素－Ｆｅ－Ｂ系薄膜永久磁石を提供する。

【構成】 Ｒ（Ｒは希土類元素のうちＮｄおよび／またはＰｒを表わす）とＦｅおよびＢを主成分とする合金からなる希土類元素－Ｆｅ－Ｂ系薄膜永久磁石であって、非磁性相である粒界が膜厚方向に貫いて形成されると共に、粒子相互間で磁気的に分離された磁気的主相が膜厚方向に延びる柱状構造を有して形成されたものであり、且つ該柱状構造相のアスペクト比（長さ／直径比）が５以上である。



【特許請求の範囲】

【請求項１】　Ｒ（Ｒは希土類元素のうちＮｄおよび／またはＰｒを表わす）とＦｅおよびＢを主成分とする合金からなる希土類元素－Ｆｅ－Ｂ系薄膜永久磁石であって、非磁性相である粒界が膜厚方向に貫いて形成されると共に、粒子相互間で磁気的に分離された磁気的主相が膜厚方向に延びる柱状構造を有して形成されたものであり、且つ該柱状構造相のアスペクト比（長さ／直径比）が５以上であることを特徴とする希土類元素－Ｆｅ－Ｂ系薄膜永久磁石。

【請求項２】　Ｒ：８～１８原子%、Ｂ：５～１０原子%、残部：実質的にＦｅからなるものである請求項１に記載の希土類元素－Ｆｅ－Ｂ系薄膜永久磁石。

【請求項３】　Ｆｅの一部を１．５原子%以下のＡｇで置換したものである請求項２に記載の希土類元素－Ｆｅ－Ｂ系薄膜永久磁石。

【発明の詳細な説明】

【０００１】

【産業上の利用分野】本発明は、優れた磁気特性を有する希土類元素－Ｆｅ－Ｂ系薄膜永久磁石に関し、この磁石は一般家庭用の各種電気製品やコンピュータを始めとし、高性能小型モータ等の電気・電子材料の素材として有用である。

【０００２】

【従来の技術】永久磁石とは、外部から電気的エネルギーを供給しなくとも磁界を発生する材料であり、その需要は上記の様な電気・電子材料を主体にしてますます増大してきている。近年、フェライト磁石およびアルニコ磁石に次ぐ第３の永久磁石として、希土類磁石が注目を集めている。この希土類磁石は電気製品や精密機器類の小型化および高精度化に寄与し得る優れた磁気的性質を発揮するものと期待され、物性研究面および生産技術面共に活発な進展を見せている。中でも近年特に期待をされているのは、希土類元素－遷移元素－Ｂ系、例えばＮｄ－Ｆｅ－ＢやＰｒ－Ｆｅ－Ｂ等の永久磁石であり、この系の永久磁石は高い保磁力（ｉＨｃ）を有すると共に、残留磁束密度（Ｂｒ）と保磁力（ｉＨｃ）の積で示される最大エネルギー積［以下（ＢＨ）$_{max}$　で表わすことがある］も高いと言われている。

【０００３】本発明の永久磁石組成は、希土類元素（但し、Ｎｄおよび／またはＰｒ）－Ｆｅ－Ｂを基本成分とする他、必要によって第４の成分としてＡｇを含むものであって、その評価については追って詳述するが、以下の説明においては便宜上、希土類元素－Ｆｅ－Ｂ系（以下、Ｒ－Ｆｅ－Ｂ系と略称することがある）の３元素磁石を代表的に取り上げて述べることとする。

【０００４】ところでＲ－Ｆｅ－Ｂ系磁石の製造方法としては、焼結法や液体急冷法等が代表的な方法として知られており、これらの方法によって薄膜永久磁石の形成が行なわれている。そして磁石材料の磁気特性は、微細組織に大きく影響されることが知られており、例えば焼結型磁石では、図２に示す様に結晶粒界相１が主相結晶粒２を十分に取り囲む構造とすることによって（尚図中３は基板を示す）、逆磁区の発生サイトを減少させて磁気特性を向上させることができると言われている（日本金属学会誌，第５７巻第４号（１９９３）Ｐ４７０～４７７）。また高速急冷型磁石では、微細な主相結晶粒と非晶質から構成され、急冷速度を調整することによって高い磁気特性が得られると言われている（電気学会論文誌Ａ，１１３巻第４号（１９９３），Ｐ２５１～２６０）。

【０００５】

【発明が解決しようとする課題】しかしながら、上記した様な焼結法や液体急冷却によって得られたＲ－Ｆｅ－Ｂ系永久磁石では、膜厚方向で見ると結晶粒は粒界相を挟んで不規則に積み重なっている構造であり、垂直磁気異方性に対する形状磁気異方性の寄与は小さい。また保磁力は、逆磁区の発生磁界に左右される逆磁区発生型であり、実際の保磁力は、理論的に一軸異方性を有する単磁区粒子の磁化反転磁界Ｈａの１／１０～２／５程度と小さいものである。こうしたことから、上記いずれの方法によっても、薄肉化と高性能化という両特性を満足することのできる永久磁石は実現できず、小型化，高性能化という近年の要求に十分応えることのできる製品用の永久磁石の実現が望まれているのが実情である。

【０００６】一方、Ｒ－Ｆｅ－Ｂ系薄膜永久磁石の新しい形成方法として、例えば特開平４－９９０１０号に見られる様なスパッタリング法による方法も提案されている。この技術は、主相である$R_2Fe_{14}B$相の磁化容易軸であるＣ軸を膜厚方向に成長させ、膜厚方向に強い異方性を有する膜を形成するものである。しかしながら、この技術では、非磁性の粒界相や形状磁気異方性については何ら考慮されておらず、磁気特性の更なる向上が望まれる。

【０００７】本発明はこうした状況の下になされたものであって、その目的は、保磁力機構を従来の逆磁区発生型から回転磁化型となるような粒界相が、粒子間を磁気的に分離すると共に、形状磁気異方性の効果が付与される様な膜を実現し、薄肉であってしかも良好な磁気特性を発揮することのできる希土類元素－Ｆｅ－Ｂ系薄膜永久磁石を提供することにある。

【０００８】

【課題を解決するための手段】上記目的を達成し得た本発明とは、Ｒ（Ｒは希土類元素のうちＮｄおよび／またはＰｒを表わす）とＦｅおよびＢを主成分とする合金からなる希土類元素－Ｆｅ－Ｂ系薄膜永久磁石であって、非磁性相である粒界が膜厚方向に貫いて形成されると共に、粒子相互間で磁気的に分離された磁気的主相が膜厚方向に延びる柱状構造を有して形成されたものであり、且つ該柱状構造相のアスペクト比（長さ／直径比）が５

以上である点に要旨を有する希土類元素－Ｆｅ－Ｂ系薄膜永久磁石である。本発明の希土類元素－Ｆｅ－Ｂ系薄膜永久磁石はＲ：８～１８原子％、Ｂ：５～１０原子％、残部：実質的にＦｅからなる化学成分組成のものが好ましく、またＦｅの一部を１．５原子％以下までの範囲でＡｇで置換することも有効である。

【０００９】

【作用】本発明者らは、これまで提案されてきた技術を基礎とし、薄肉でしかも高性能な永久磁石を実現すべく、特にその結晶組織構造と磁気特性の関係について検討を重ねた。その結果、非磁性相である粒界相と磁気的主相である$R_2Fe_{14}B$相が特定の組織構造を呈しているものでは、薄肉であっても極めて優れた磁気特性を示すことがわかった。即ち、図１に示す様に、非磁性相である粒界４が膜厚方向に貫いて形成されると共に、粒子相互間で分離された磁気的主相５（例えば$R_2Fe_{14}B$相）が膜厚方向に延びる柱状構造を有して形成されたものであり、且つ該柱状構造相のアスペクト比（長さ／直径比）が所定の範囲のものでは（尚図１中、３は基板を示す）、従来技術で示した様な永久磁石に比べて、$(BH)_{max}$ が顕しく向上することを見出し、本発明を完成した。

【００１０】本発明によって上記の様な効果が得られた理由については、その全てを解明し得た訳ではないが、おそらく次の様に考えることができる。即ち、上記の様な組織構造では、磁気的主相がアスペクト比が大きい柱状構造を有しているので、結晶磁気異方性以外に形状磁気異方性が生じて膜厚方向の磁気異方性が大きくなると共に、前記磁気的主相が粒子相互間で磁気的に分離されているので粒子間の磁気的相互作用が弱まって、保磁力が大きくなるものと考えられる。

【００１１】次に、本発明のＲ－Ｆｅ－Ｂ系磁石を構成する合金組成について説明する。まず希土類元素としては、Ｎｄおよび／またはＰｒとする必要がある。即ち、最も高い磁気特性が得られるのは、Ｐｒ，Ｎｄであるので、希土類元素としてはＰｒおよび／またはＮｄであることが必要であり、他の希土類元素を含んでいると、希望する磁気特性が得られない。

【００１２】本発明のＲ－Ｆｅ－Ｂ系磁石において、Ｒが少な過ぎると主相である$R_2-Fe_{14}-B$（原子比、例えば$Pr_2Fe_{14}B$）を形成されにくくなり、磁石の高性能化は達成されない。こうした観点からして、Ｒの割合は８原子％以上とするのが好ましい。一方上限については、１８原子％を超えると、非磁性相であるＲリッチ相の過剰を招きこれが磁束密度（Ｂｒ）の低下等となって現われ、良好な磁気特性を発揮することはできない。尚Ｒのより好ましい組成割合は、１２～１５原子％の範囲である。

【００１３】一方、Ｂは５～１０原子％とするのが好ましく、５原子％未満では主相体積率の不足が生じ、磁束密度（Ｂｒ）の低下を招く。他方上限については、磁気特性を有しない$R_2Fe_4B_4$相の出現による保磁力（ｉＨｃ）の低下を防止するという観点から１０原子％以下とするのが好ましい。尚Ｂのより好ましい範囲は、７～９原子％程度である。

【００１４】本発明のＲ－Ｆｅ－Ｂ系磁石において、上記ＲとＢ以外は実質的にＦｅ（即ち、Ｆｅおよび不可避不純物）であるが、Ｆｅの一部をＡｇで置換することも有効である。即ち、Ａｇを添加すると粒子間の磁気的孤立度が向上し、$(BH)_{max}$ が増大する。但し、Ａｇの添加量を余り多くすると非磁性の粒界相が増加し、$(BH)_{max}$ が却って低下するので、１．５原子％以下とすべきである。尚Ａｇの添加量のより好ましい範囲は１原子％以下である。

【００１５】本発明のＲ－Ｆｅ－Ｂ系薄膜磁石における前記柱状構造相（磁気的主相）のアスペクト比（長さ／直径比）は、５以上とする必要があるが、この値が５未満では形状磁気異方性付与による特性の向上が小さいものとなる。

【００１６】尚本発明者らは、アスペクト比、$(BH)_{max}$ および膜厚の関係について検討したところ、下記の点について確認している。即ち、Ｎｄ－Ｆｅ－Ｂ系薄膜磁石では、膜厚が５μｍでアスペクト比が５～１５のもので$(BH)_{max}$ が１７～１８ＭＧＯｅが達成されており、膜厚２０μｍでアスペクト比が２０～５０のもので$(BH)_{max}$ が１４～１６ＭＧＯｅが達成されていた。

【００１７】本発明のＲ－Ｆｅ－Ｂ系薄膜永久磁石を製造する方法としては、基本的にスパッタリング法によって柱状構造を有する磁気的主相を形成する様にすれば良いが、上記の様な微細組織を形成することができれば、真空蒸着法やＣＶＤ法等も採用することもできる。また製造する際の条件については特に限定するものではないが、上記組織を形成するためには、特に製造時の基板温度を５００～７００℃程度とするのが好ましい。即ち、このときの基板温度が５００℃未満では、膜がアモルファス状態になって硬磁性が得られず、７００℃を超えると$R_2Fe_{14}B$以外の相（例えばα－Ｆｅ相等）が生成して、$(BH)_{max}$ が減少することになる。更に、本発明のＲ－Ｆｅ－Ｂ系薄膜永久磁石を製造する際のスパッタガス圧は、従来方法（例えば8×10^{-3} Ｔｏｒｒ程度）よりも高くするのが良い（例えば15×10^{-3} Ｔｏｒｒ程度）。

【００１８】以下本発明を実施例によって更に詳細に説明するが、下記実施例は本発明を限定する性質のものではなく、前・後記の趣旨に徴して設計変更することはいずれも本発明の技術的範囲に含まれるものである。

【００１９】

【実施例】

実施例１

組成割合を変えた合金ターゲットを使用して、ＤＣスパ

ッタリング法（スパッタガス圧：１５×１０$^{-3}$ Ｔｏｒｒ）により様々な組成割合のＮｄ－Ｆｅ－Ｂ合金薄膜磁石を作成した。このとき基板温度は、６００℃とした。また薄膜の膜厚は２０μｍであった。更に、得られた薄膜は、前記図１に示した結晶組織であることを、電子顕微鏡観察によって確認した。得られた合金薄膜磁石について、各々振動試料型磁力計（ＶＳＭ）によって、最大エネルギー積（ＢＨ）$_{max}$ を測定した。図３には、ＮｄとＢとＦｅを基本組成とする薄膜磁石の最大エネルギー積（ＢＨ）$_{max}$ の値が１４ＭＧＯｅを超える組成範囲を斜線で示す。尚１４ＭＧＯｅを超えるものは、アスペクト比が２０～５０であった。

【００２０】実施例２

組成割合を変えた合金ターゲットを使用して、ＤＣスパッタリング法（スパッタガス圧：１５×１０$^{-3}$ Ｔｏｒｒ）により様々な組成割合のＰｒ－Ｆｅ－Ｂ合金薄膜磁石を作成した。このとき基板温度は、６００℃とした。また薄膜の膜厚は２０μｍであった。更に、得られた薄膜は、前記図１に示した結晶組織であることを、電子顕微鏡観察によって確認した。得られた合金薄膜磁石について、各々ＶＳＭによって、（ＢＨ）$_{max}$ を測定した。図４には、ＰｒとＢとＦｅを基本組成とする薄膜磁石の（ＢＨ）$_{max}$ の値が１４ＭＧＯｅを超える組成範囲を斜線で示す。尚１４ＭＧＯｅを超えるものは、アスペクト比が２０～５０であった。

【００２１】実施例３

ＮｄとＢの組成を一定にして、残部のＦｅとＡｇの組成を変化させて成膜した。図５は、$Nd_{12}-Fe_{83-x}-B_{5}-Ag_{x}$ 膜のＡｇ添加量（ｘ）の変化による（ＢＨ）$_{max}$ の変化を示すグラフである。尚図５の結果は、それぞれの組成において、最大値が得られた成膜条件で行なったものである。また得られた合金薄膜磁石は、アスペクト比が２０～５０で図１に示した結晶組織であることを電子顕微鏡観察によって確認した。この結果から明らかな様に、１．５原子％以下のＡｇの添加は（ＢＨ）$_{max}$ の向上に有効であることがわかる。

【００２２】実施例４

ＰｒとＢの組成を一定にして、残部のＦｅとＡｇの組成を変化させて成膜した。図６は、$Pr_{12}-Fe_{83-x}-B_{5}-Ag_{x}$ 膜のＡｇ添加量（ｘ）の変化による（ＢＨ）$_{max}$ の変化を示すグラフである。尚図６の結果は、それぞれの組成において、最大値が得られた成膜条件で行なったものである。また得られた合金薄膜磁石は、アスペクト比が２０～５０で図１に示した結晶組織であることを電子顕微鏡観察によって確認した。この結果から明らかな様に、Ｐｒ－Ｆｅ－Ｂ系においても１．５原子％以下のＡｇの添加は（ＢＨ）$_{max}$ の向上に有効であることがわかる。

【００２３】比較例

１５．０原子％Ｐｒ－７９．２原子％Ｆｅ－５．５原子％Ｂ－０．３原子％Ａｇの合金を、アルゴンガス雰囲気中で高周波加熱より溶解し、ロール表面速度１０ｍ／ｓで回転する鋼製の単ロール（２００ｍｍφ）上に噴出し急冷薄帯を作成した。このときの膜厚は２０μｍであった。また得られた薄膜は、前記図２に示した結晶組織であることを、電子顕微鏡観察によって確認した。得られた薄膜について、ＶＳＭによって、（ＢＨ）$_{max}$ を測定したところ、最大で１４ＭＧＯｅしか得られなかった。

【００２４】

【発明の効果】本発明は以上の様に構成されており、薄肉であってしかも良好な磁気特性を発揮することのできる希土類元素－Ｆｅ－Ｂ系薄膜永久磁石が実現できた。

【図面の簡単な説明】

【図１】本発明に係る永久磁石の微細組織を示す模式図である。

【図２】従来の永久磁石の微細組織を示す模式図である。

【図３】本発明のＮｄ－Ｆｅ－Ｂ系薄膜磁石の（ＢＨ）$_{max}$ の値が１４ＭＧＯｅを超える組成範囲を示したグラフである。

【図４】本発明のＰｒ－Ｆｅ－Ｂ系薄膜磁石の（ＢＨ）$_{max}$ の値が１４ＭＧＯｅを超える組成範囲を示したグラフである。

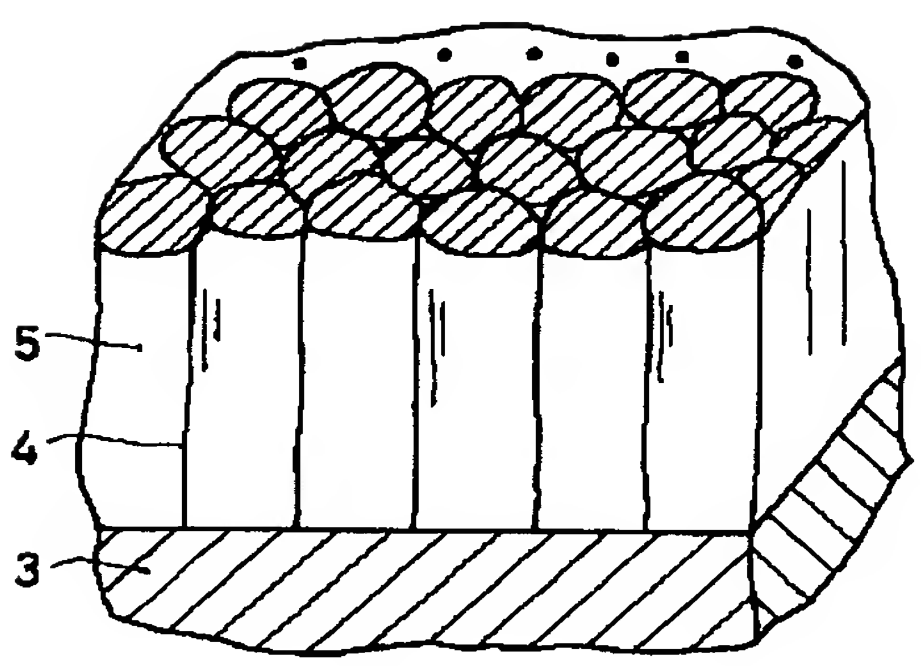
【図５】本発明の$Nd_{12}-Fe_{83-x}-B_{5}-Ag_{x}$薄膜磁石の（ＢＨ）$_{max}$ におけるＡｇ添加量による変化を示したグラフである。

【図６】本発明の$Pr_{12}-Fe_{83-x}-B_{5}-Ag_{x}$薄膜磁石の（ＢＨ）$_{max}$ におけるＡｇ添加量による変化を示したグラフである。

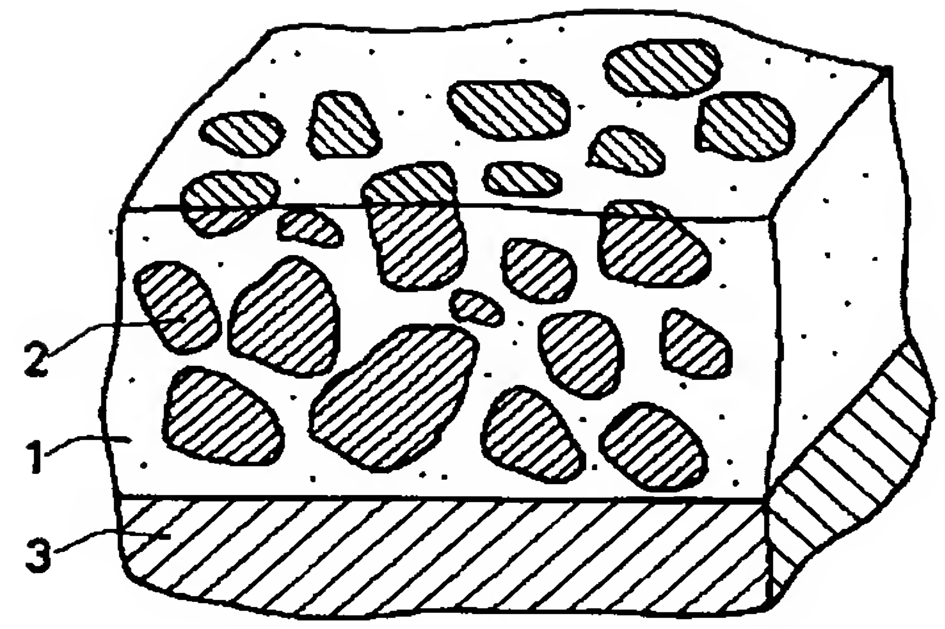
【符号の説明】

１　結晶粒界相
２　主相結晶粒
３　基板
４　粒界
５　磁気的主相

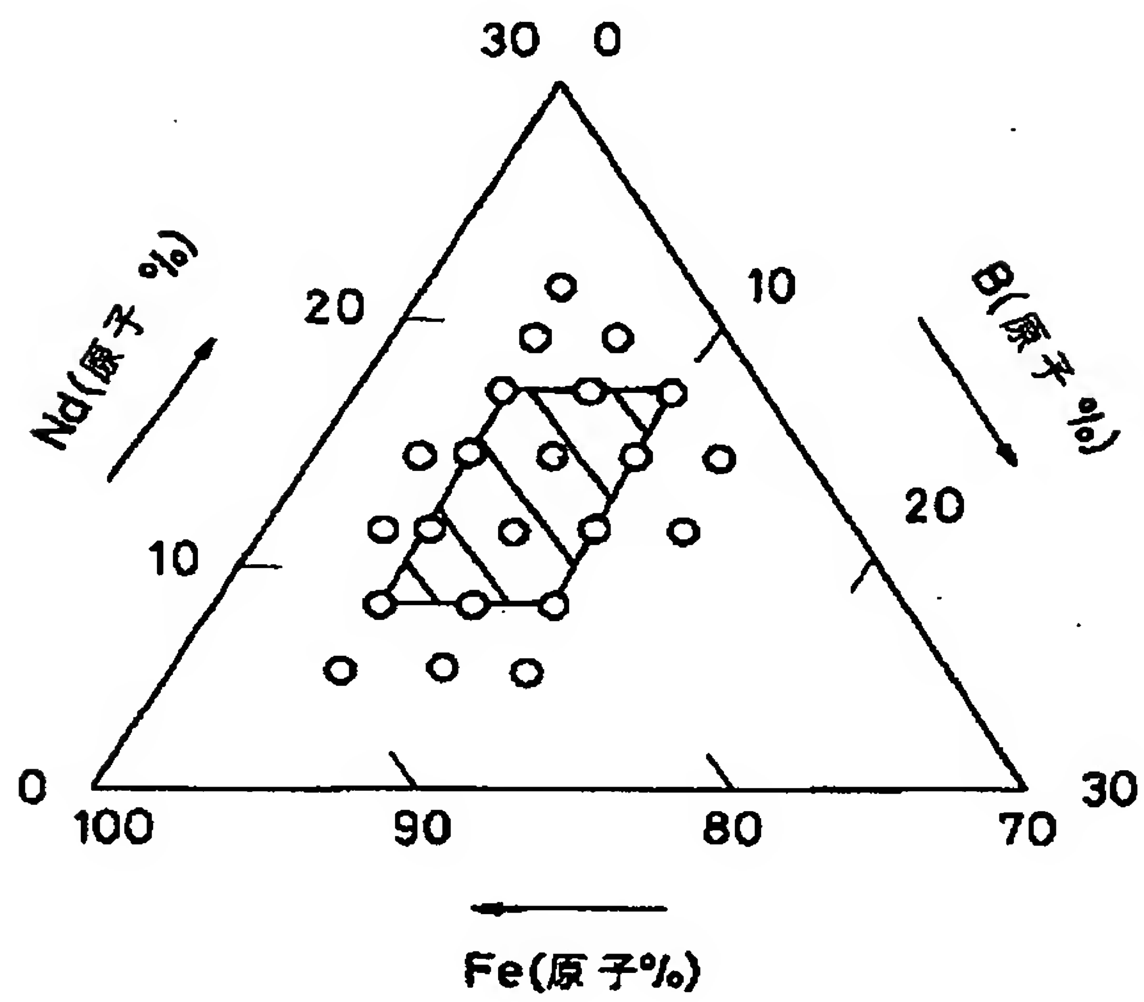
【図１】



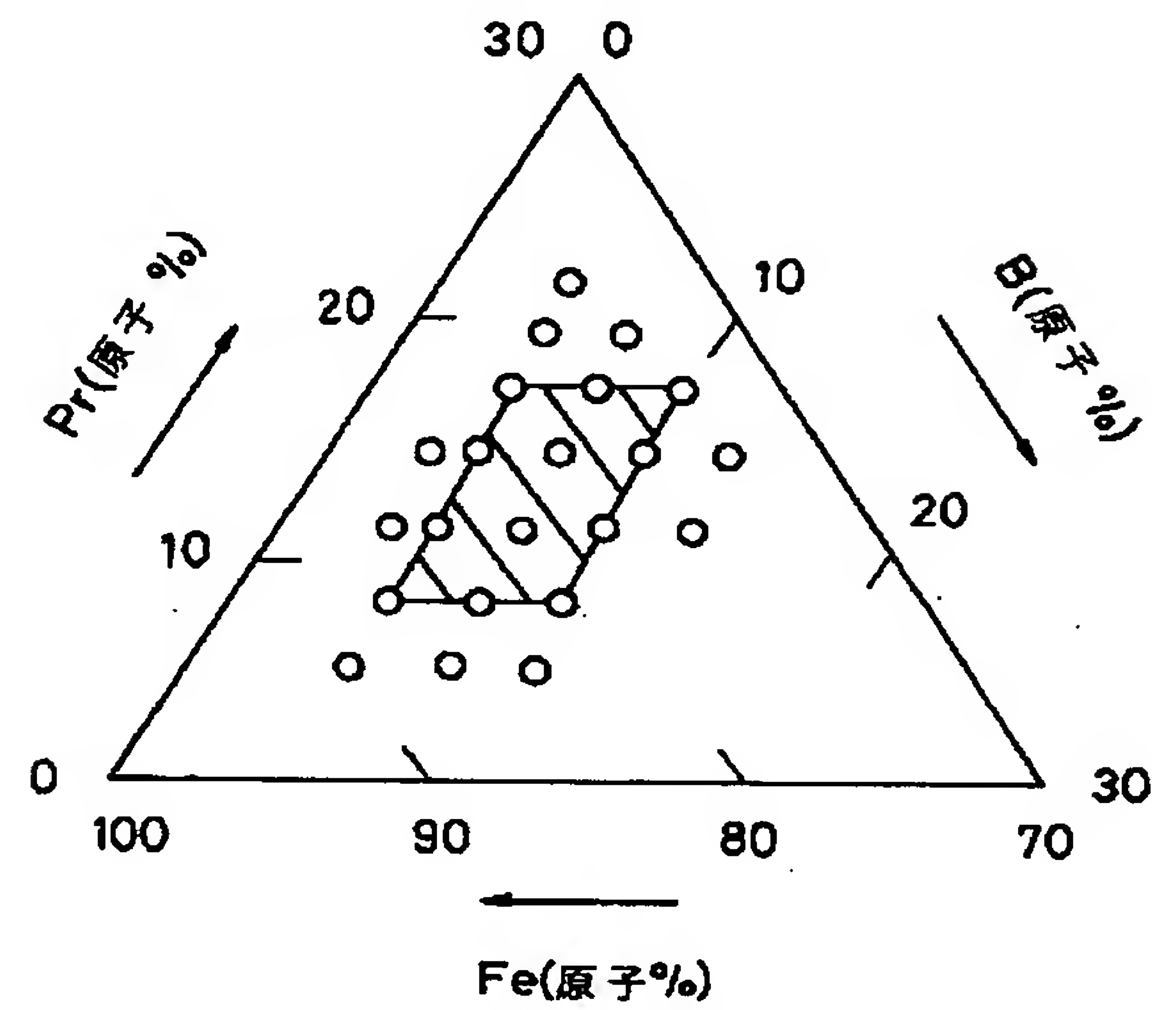
【図２】



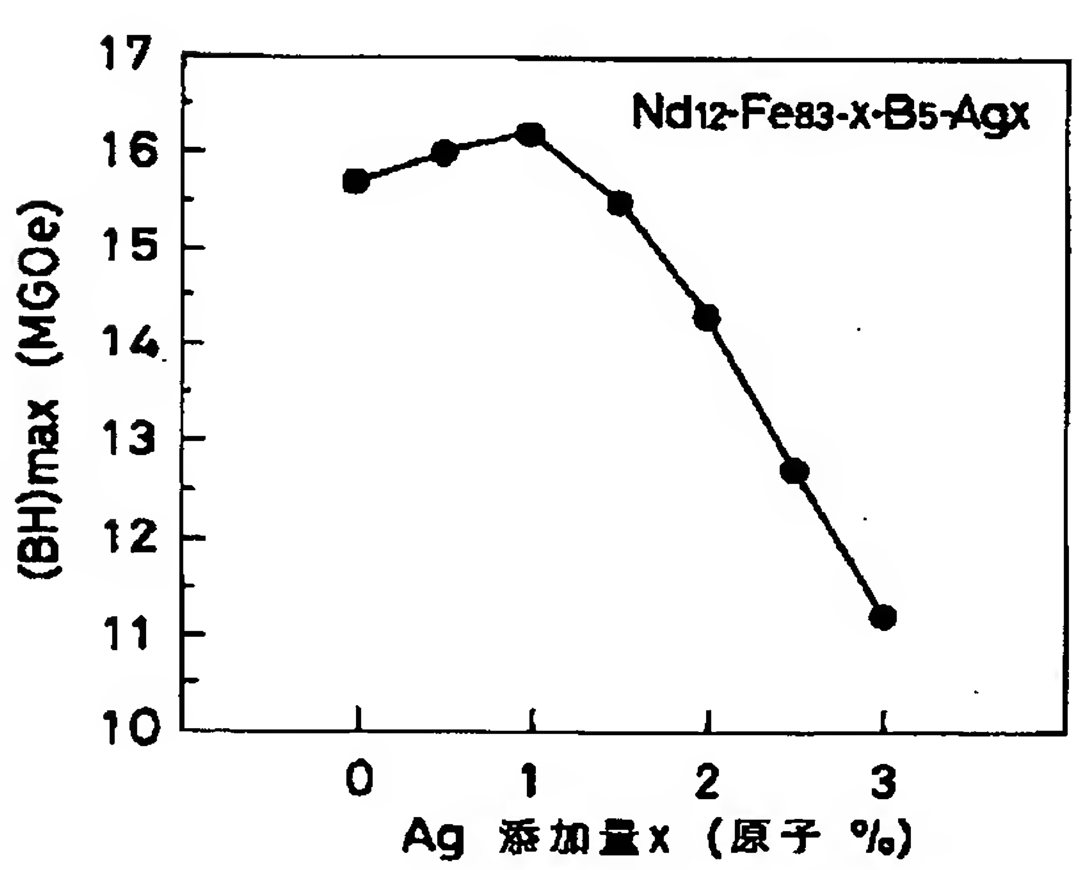
【図３】



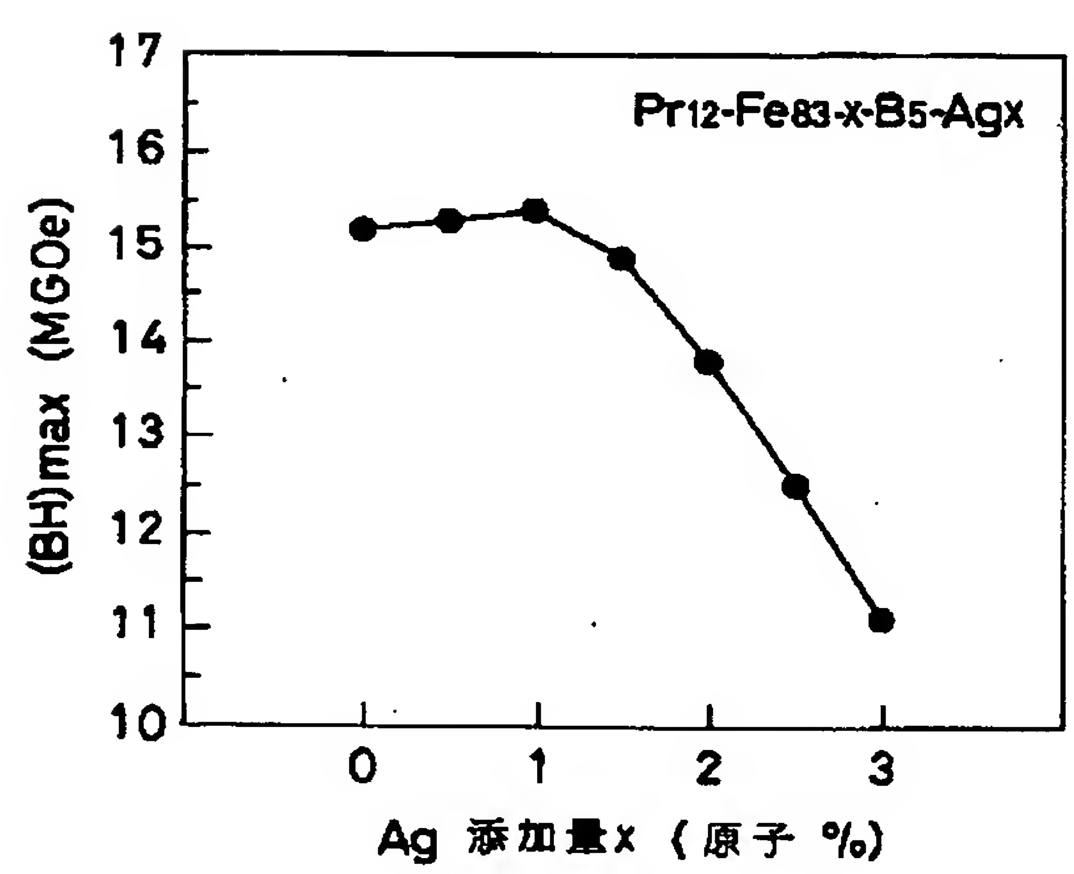
【図４】



【図５】



【図６】



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